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ABSTRACT

The fractional independent yield of ^{140}La from the thermal-neutron induced fission of ^{249}Cf has been measured by two methods. In one method, the change in activity as a function of time of the 1596.2-keV gamma ray from ^{140}La was measured with a Ge(Li) detector in the presence of the gross fission products. In the other method, ^{140}La was chemically separated from ^{140}Ba at known times after fission and its gamma activity was measured. The weighted mean of the yields from the two methods was 0.035 ±0.001.

This yield is consistent with other yields from ^{249}Cf and ^{252}Cf fission which show there may be less redistribution of charge during fission of ^{249}Cf and ^{252}Cf than during fission of ^{233}U and ^{235}U. The most probable charge, Z_p, for A = 134, 136, 138, and 140 appears to be 0.3 to 0.4 charge units closer to unchanged charge distribution for fission of ^{249}Cf and ^{252}Cf than for fission of ^{233}U and ^{235}U.

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INTRODUCTION

A series of experiments is underway in this laboratory to measure fractional cumulative and independent yields of products of thermal-neutron induced fission of ^{249}Cf [1]. The first experiments in this series were restricted to fission product decay chains such as A = 140 (Fig. 1) in which the member is partially shielded by a long-lived precursor. The 40.2-hr ^{140}La is shielded from decay of its short-lived precursors by 12.8-d ^{140}Ba so that only small amounts of ^{140}La are formed by beta decay the first few hours after fission.

Fractional independent yields of ^{140}La from thermal-neutron fission of ^{233}U and ^{235}U were reported by Grummitt and Milton [2]. Fractional cumulative yields of ^{140}Xe from ^{235}U fission were reported by Wahl [3] and from ^{233}U fission by Wolfsberg [4]. Wahl *et al.* [5] also reported the fractional independent yield of ^{140}Ba from ^{235}U fission, and Eichor and Troutner [6] reported the ^{140}Ba yield from ^{233}U fission. From these last results, it is possible to calculate fractional cumulative yields of ^{140}Cs. The A = 140 decay chain is, therefore, one of the few chains for which three or more fractional cumulative yields have been measured for ^{235}U fission, and the only one for which three fractional cumulative yields have been reported for ^{233}U fission.

All of the yields above, except that for ^{140}La from ^{235}U fission, are in general agreement with the description of charge distribution proposed by Wahl *et al.* [7]. In that description, charge distribution is represented by a Gaussian function with a standard deviation, σ, of 0.56 ±0.06 and a maximum, Z_p, given by

$$(Z_p)_H = [A'_H\ (Z_F/A_F) - (0.45 \pm 0.10)] \qquad (1)$$

where A_H' is the heavy fragment mass-number before neutron emission; Z_F is the charge of the fissioning nucleus; and A_F is the mass-number of the fissioning nucleus. The quantity A_H' (Z_F/A_F) represents the charge of nucleus of mass A_H' for which the Z/A ratio is the same as that of the fissioning nucleus and will be referred to as UCD, for unchanged charge distribution.

There is an odd-even effect [7] which tends to make yields of even-Z fragments greater than predicted by the relation above. Other evidence indicates a shell effect which results in somewhat lower yields of 81- and 83-neutron nuclides and somewhat higher yields for 82-neutron nuclides.

Recent work [1] from this laboratory, however, showed that fractional cumulative yields of ^{132}Te and ^{134}Te from thermal-neutron induced fission of ^{249}Cf were lower than predicted by the Wahl *et al.* [7] prescription and much lower if the odd-even effect was considered. There is also some evidence that the fractional cumulative yield of ^{136}Xe (calculated from the independent yield of ^{136}Cs reported by Flynn and von Gunten [8]) for that fission process is lower than predicted by the Wahl *et al.* [7] relation. We have measured the fractional independent yield of ^{140}La from ^{249}Cf fission to learn whether it is more consistent with other yields from ^{249}Cf fission or with those from ^{233}U and ^{235}U fission.

EXPERIMENTAL

Two experiments were performed to measure the ^{140}La independent yield. In Procedure A, no chemical separations were done and the change in activity as a function of time of the 1596.2-keV gamma ray from ^{140}La was measured. In Procedure B, ^{140}La was chemically separated from ^{140}Ba at known times after fission, and its gamma activity was measured.

Irradiations

Samples were prepared for irradiation by electroplating ∿1 μg of ^{249}Cf on high purity aluminum foils. The foils were covered with other aluminum foils to serve as catcher foils, sealed in quartz, and irradiated for two minutes at a flux greater than 10^{15} neutrons/cm^2/sec. In Procedure A, the source foil was placed in a 2-dram vial that was sealed with plastic tape. In Procedure B, the catcher foil was removed and dissolved as described below.

Chemical Procedures

For Procedure B, the catcher foil was dissolved in ∿6M HCl containing ∿160 mg each of Ba(II) and La(III) carriers in a total volume of ∿200 ml. Five ∿20-ml portions were used for the experiment. Lanthanum was separated from barium in each portion by adding NaOH to precipitate $La(OH)_3$. The mixture was centrifuged, and the $La(OH)_3$ dissolved in HCl and reprecipitated with NaOH. Following centrifugation, this precipitate was redissolved, and two more such precipitations were made with NH_4OH. The final precipitate was dissolved in ∿10 ml of 1M HCl and a 5-ml aliquot pipetted into a 2-dram counting vial.

The time at which the first precipitation occurred was taken as the time of separation of lanthanum from barium. Three separations were done during the first few hours after fission and two more several days later. After the gamma activity measurements were completed, the amount of lanthanum in each vial was measured by titrating with EDTA [9] to determine the chemical yield.

Counting

The gamma radioactivity of each sample was measured with a 15-cm^3 Nuclear Diodes trapezoidal coaxial Ge(Li) detector and a 4096-channel

pulse height analyzer. For the 1332.5 keV ^{60}Co gamma ray, the system-contributed resolution was 3.2 keV (FWHM).

In Procedure A, the sample holder was designed to reproduce the geometry from one count to another. The sample was 4 in. from the face of the detector. An absorber package consisting of 1/2-in. lead absorber and a 1/2-in. "Plexiglas"* absorber was placed between the sample and the detector. For Procedure B the total activities were much smaller, and the samples were positioned flush against the face of the detector.

The spectrum from each count was analyzed on an IBM-360/65 computer using SPAN [10], a program which measures the area under each peak. In Procedure A, there was another peak near 1596 keV, probably due to ^{112}Ag in equilibrium with ^{112}Pd. To ensure against error in the background as estimated by the computer, the ^{140}La peaks for each run were plotted by hand, the background estimated visually, and the area calculated. The hand integrated areas used in the calculations agreed with the computer results. For Procedure B, there were no other peaks in the 1596 keV energy region, and the computer results were used directly.

Calculations

Because the half-lives of ^{140}I, ^{140}Xe, and ^{140}Cs are very short compared to times of separation or counting, the mass-140 decay chain will be assumed to consist only of ^{140}Ba, ^{140}La, and the stable ^{140}Ce. Also, the fractional independent yield of ^{140}Ce will be assumed to be very small compared to that of ^{140}Ba or ^{140}La.

For Procedure A, the number of atoms of ^{140}La that have decayed during any count is equal to the number of atoms of ^{140}Ce produced during that

* Trademark of Rohm and Haas Company

time. The number of counts observed for ^{140}La is of course the number of disintegrations multiplied by both the counting efficiency and the fractional live time of the detection system.

These relations can be summarized as:

$$\frac{A_{La}t' \, e^{\lambda_{La}t}}{t'' \, (1-e^{-\lambda_{La}t}} = \left[\frac{\lambda_{La} \, e^{\lambda_{La}t} \, (e^{-\lambda_{Ba}t'}-1)}{(\lambda_{Ba}-\lambda_{La}) \, e^{\lambda_{Ba}t} \, (1-e^{-\lambda_{La}t'})} + \frac{\lambda_{Ba}}{(\lambda_{Ba}-\lambda_{La})} \right] \varepsilon N^o_{Ba} + \varepsilon N^o_{La} \quad (2)$$

where A_{La} is the observed counts of ^{140}La; ε, counting efficiency for ^{140}La; t'', live counting time; t', elapsed counting time; t, time interval between fission and start of count; $\lambda_{Ba}, \lambda_{La}$, decay constants for ^{140}Ba and ^{140}La, respectively; and N^o_{Ba}, N^o_{La}, number of atoms of ^{140}Ba and ^{140}La, respectively, at time of fission.

The above equation can be rewritten as

$$Y = \varepsilon \, X \, N^o_{Ba} + \varepsilon N^o_{La} \quad (3)$$

where Y is the left side of Equation (2) and X is the coefficient of εN^o_{Ba} in Equation (2). Note that counting conditions were held constant so that ε is a constant. For counts begun at times much longer than the half-life of ^{140}La, Y is much larger than εN^o_{La}, so that εN^o_{Ba} can be calculated directly for those counts. The average value of εN^o_{Ba} can then be used to calculate εN^o_{La} for the counts at earlier times.

Because the fractional independent yield of ^{140}La is given by $N^o_{La}/(N^o_{Ba} + N^o_{La})$, it must therefore be equal to $\varepsilon N^o_{La}/(\varepsilon N^o_{Ba} + \varepsilon N^o_{La})$.

For Procedure B, a similar equation can be used.

$$\frac{A_{La}}{c} = \frac{\varepsilon \lambda_{La}}{c} \left[\frac{\lambda_{Ba}(e^{-\lambda_{Ba}T}-e^{-\lambda_{La}T})N^o_{Ba}}{(\lambda_{La}-\lambda_{Ba})} + N^o_{La} \, e^{-\lambda_{La}T} \right] e^{-\lambda_{La}(t-T)} (1-e^{-\lambda_{La}t'}) \frac{t''}{t'} \quad (4)$$

Symbols are the same as in earlier equations with the addition of T, which is the time interval between fission and the lanthanum-barium separation and c, the concentration of lanthanum in each sample. This term is necessary to normalize all samples to a constant chemical yield.

Equation (4) can be rearranged to

$$\frac{A_{La}\, e^{\lambda_{La} t}\, t'}{c\,(1-e^{-\lambda_{La} t'})\, t''} = \varepsilon\lambda_{La}\left[\frac{\lambda_{Ba}(e^{-(\lambda_{Ba}-\lambda_{La})T}-1)}{(\lambda_{La}-\lambda_{Ba})}\,\frac{N^{o}_{Ba}}{c} + \frac{N^{o}_{La}}{c}\right] \tag{5}$$

An equation of the form

$$Y' = k\left[X'\,\frac{N^{o}_{Ba}}{c} + \frac{N^{o}_{La}}{c}\right] \tag{6}$$

can be written to represent Equation 5.

Y' and X' have the same meaning as Y and X in Equation (3) but have values consistent with Equation (5). The symbol k represents $\varepsilon\lambda_{La}$.

The fractional independent yield of ^{140}La is

$$\frac{kN^{o}_{La}}{c}\Bigg/\left(\frac{kN^{o}_{Ba}}{c} + \frac{kN^{o}_{La}}{c}\right)$$

RESULTS

Data for both procedures are shown in Tables I and II. Uncertainties shown for values of A_{La} are standard deviations expected on the basis of Poisson statistics for peak areas and backgrounds resulting from computer integration of the peaks.

Values of X, Y, X', Y', and the values of εN^{o}_{La}, εN^{o}_{Ba}, kN^{o}_{La}, and kN^{o}_{Ba} calculated from them are shown in Tables III and IV. Uncertainties for Y represent the uncertainties in peak areas and also in chemical yields for

Procedure B. Uncertainties in average values are standard deviations of means. Because the uncertainties for different values of the same quantity, i.e., the four values for ϵN^{o}_{La}, are not greatly different, no weighting has been used in calculating the averages. From these values, the calculated fractional independent yield of ^{140}La is 0.0349 ±0.0012 from the results of Procedure A and 0.0374 ±0.0040 from the results of Procedure B. The weighted mean rounded to the nearest 0.001 is 0.035 ±0.001.

The half-lives of ^{140}Ba and ^{140}La are well known. Times were measured to the nearest minute so that uncertainties in times of separation and counting are small. There are no other significant uncertainties to be added to the value above.

DISCUSSION

One of the characteristics of a Gaussian charge distribution for a given mass chain is that fractional cumulative yields fall on a straight line when plotted as a function of Z on probability paper. The slope of the line is a measure of σ, and the line passes through a probability of 0.5 at $Z = Z_p - 0.5$. Yields for different mass chains and even for different fission processes can be shown on the same plot, if they are plotted as a function of $Z-Z_p$. If Z_p is given by Equation (1), Z_H-Z_p is equivalent to Z_H-UCD + 0.45. Yields can therefore be plotted as a function of Z_H-UCD. If this is done, the line passes a probability of 0.50 at Z_H-UCD = -0.95. However, it has been shown [6,7,11] that in the A = 140 mass region, there are odd-even effects which result in enhancement of yields of even-Z products. In addition, the independent yields of 81- and 83-neutron products may be somewhat lower than predicted with a resulting increase in the fractional cumulative yields of 82- and 84-neutron nuclides. Therefore, comparison of

fractional cumulative yields from different fission processes is valid only if the yields are for the same nuclide or for nuclides of similar nuclear structure.

Fortunately, fractional cumulative yields of ^{134}Te, ^{136}Xe, ^{138}Xe, and ^{140}Ba, all of which are even-Z and 82- or 84-neutron nuclides, have been reported for both ^{233}U [4,12,13] and ^{235}U [7] fission. In addition, yields of ^{134}I [14], ^{136}Xe [15], and ^{138}Xe [14] have been reported for spontaneous fission of ^{252}Cf, and yields of ^{134}I [1] and ^{136}Xe [8] for thermal-neutron induced fission of ^{249}Cf. It is possible, therefore, to compare the fractional cumulative yield of ^{140}Ba from this work to yields of similar nuclides for several fission processes.

Fig. 2 is a probability graph showing such a comparison. Values of A' for ^{235}U fission from Wahl *et al.* [7] were rounded to the nearest 0.1. Values of A' for ^{233}U fission were estimated from the neutron emission results of Apalin *et al.* [16] to be 0.1 higher than those for ^{235}U. Values of A' for ^{252}Cf fission were estimated from the neutron emission results of Bowman *et al.* [17]. Finally A' values for ^{249}Cf fission were estimated to be $\sim$0.4 higher than for ^{252}Cf fission. This estimate is based on the results of Jaffey and Lerner [18] showing that prompt neutron emission is nearly constant for thermal-neutron induced fission of different isotopes of the same element, and on data from several sources [18-20] showing that ν for the same fissioning nucleus is 0.6 to 0.8 higher for thermal-neutron fission than for spontaneous fission. The assumption was made that this increase is equally divided between light and heavy fragments. A summary of the values of A' and yields used in constructing Fig. 2 is shown in Table V.

The line in Fig. 2 is consistent with Equation (1) and a σ of 0.56. Points for the yields from ^{233}U and ^{235}U are generally consistent with the

slope of the line but somewhat above it. Only the point for ^{140}Ba from ^{235}U fission is not consistent with the others. (Similar comparisons of yields of several isotopes of xenon and barium from ^{235}U fission [11] and yields for several members of the A = 139 and A = 140 mass chains for both ^{233}U and ^{235}U fission [6] have also shown the fractional cumulative yield of ^{140}Ba to be inconsistent with the others. It will not be considered further in this discussion.) The points for ^{252}Cf and ^{249}Cf fission, however, fall near or just below the line. The yield of ^{140}Ba from ^{249}Cf fission is consistent with other yields from ^{249}Cf fission and with those from ^{252}Cf fission, but not with those from ^{233}U and ^{235}U fission.

A similar comparison is shown in Fig. 3. Yields are the same as those in Fig. 1, but yields for fission of ^{249}Cf and ^{252}Cf have been adjusted to those from ^{233}U and ^{235}U fission in the following manner. All points for a given nuclide, i.e. ^{134}Te, are moved equal distances to the right until the points for ^{233}U and ^{235}U fission are consistent with the line. The points for ^{249}Cf and ^{252}Cf fission then fall along a line which is $\sim$0.35 charge units to the right of the line shown. If it can be assumed that nuclear structure effects are the same for all the fission processes, this indicates that the constant -0.45 in Equation (1) may be nearer to -0.10 for ^{249}Cf and ^{252}Cf fission, which is consistent with the earlier observations [1,21] that there may be less redistribution of charge during fission of ^{249}Cf and ^{252}Cf than during fission of ^{233}U and ^{235}U.

The good agreement between the ^{249}Cf and ^{252}Cf yields shown in Fig. 3 may be fortuitous. A similar analysis for yields of ^{139}Xe, ^{140}Xe, and ^{141}Xe from fission of ^{233}U [4], ^{235}U [3], and ^{252}Cf [5] shows a similar but smaller displacement. On the other hand, an analysis of yields of ^{132}Te from fission of ^{233}U [12], ^{235}U [7], and ^{249}Cf [1] shows a greater

displacement. Experiments are underway to measure other yields from ^{249}Cf fission and the same yields from fission of other transplutonium elements.

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TABLE I

Data for Procedure A

t, min	t', min	t'', min	A_{La}, counts
638	504	383	7539 ±226
1154	202	160	3729 ±119
1385	179	160	3966 ±111
1583	175	160	4136 ±116
21774	162	160	3669 ±22
22959	1223	1211	28036 ±477
34887	819	812	11876 ±118
62072	3942	3925	19259 ±154

TABLE II

Data for Procedure B

T, min	t, min	t', min	t'', min	c, mg/ml	A_{La}, counts
194	3003	409	388	0.943 ±0.004	2033 ±57
196	3418	762	725	1.18 ±0.01	3687 ±77
232	1640	335	311	1.23 ±0.01	2419 ±63
12958	13407	699	678	0.669 ±0.003	8451 ±105
12960	15831	1149	1125	0.632 ±0.004	6413 ±96

TABLE III

Results from Procedure A

Y	X	εN^{o}_{Ba}	εN^{o}_{La}
$(8.84 \pm 0.27) \times 10^{4}$	3.73×10^{-2}		$(4.14 \pm 0.14) \times 10^{4}$
$(1.16 \pm 0.37) \times 10^{5}$	5.54×10^{-2}		$(4.66 \pm 0.17) \times 10^{4}$
$(1.32 \pm 0.37) \times 10^{5}$	6.70×10^{-2}		$(4.74 \pm 0.15) \times 10^{4}$
$(1.45 \pm 0.41) \times 10^{5}$	7.79×10^{-2}		$(4.73 \pm 0.15) \times 10^{4}$
$(4.24 \pm 0.03) \times 10^{7}$	35.0	$(1.21 \pm 0.01) \times 10^{6}$	
$(6.97 \pm 0.12) \times 10^{7}$	53.6	$(1.30 \pm 0.02) \times 10^{6}$	
$(1.28 \pm 0.01) \times 10^{9}$	1.00×10^{3}	$(1.28 \pm 0.01) \times 10^{6}$	
$(1.57 \pm 0.01) \times 10^{12}$	1.25×10^{6}	$(1.26 \pm 0.01) \times 10^{6}$	
	Average	$(1.26 \pm 0.02) \times 10^{6}$	$(4.57 \pm 0.14) \times 10^{4}$

TABLE IV

Results from Procedure B

Y'	X'	kN^{o}_{Ba}	kN^{o}_{La}
$(4.85 \pm 0.14) \times 10^{4}$	7.48×10^{-3}		$(4.16 \pm 0.14) \times 10^{4}$
$(4.45 \pm 0.10) \times 10^{4}$	7.56×10^{-3}		$(3.75 \pm 0.10) \times 10^{4}$
$(3.70 \pm 0.10) \times 10^{4}$	8.98×10^{-3}		$(2.87 \pm 0.10) \times 10^{4}$
$(3.38 \pm 0.04) \times 10^{6}$	3.681	$(9.06 \pm 0.11) \times 10^{5}$	
$(3.49 \pm 0.05) \times 10^{6}$	3.684	$(9.36 \pm 0.14) \times 10^{5}$	
	Average	$(9.21 \pm 0.15) \times 10^{5}$	$(3.59 \pm 0.38) \times 10^{4}$

TABLE V

A Comparison of Fractional Cumulative Yields of ^{134}Te, ^{136}Xe, ^{138}Xe, and ^{140}Ba from Fission of ^{233}U, ^{235}U, ^{249}Cf, and ^{252}Cf

A	^{233}U		^{235}U		^{249}Cf		^{252}Cf	
	A'[a]	Yield	A'[a]	Yield	A'[a]	Yield	A'[a]	Yield
134	135.0	0.62 ±0.03[b]	134.9	0.89 ±0.01[e]	135.5	0.29 ±0.06[f]	135.1	0.74 ±0.03[i]
136	137.2	0.986 ±0.004[c]	137.1	0.99903 ±0.00005[e]	137.8	0.945 ±0.011[g]	137.4	0.9952 ±0.0010[j]
138	139.2	0.827 ±0.012[c]	139.1	0.953 ±0.002[e]	140.0	—	139.6	0.89 ±0.03[i]
140	141.3	0.9962 ±0.0001[d]	141.2	0.9993 ±0.0001[d]	142.1	0.964 ±0.001[h]	141.7	—

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a. Estimation of A' values described in text.
b. Ref. 12
c. Ref. 4
d. Ref. 2
e. Ref. 5
f. Ref. 1
g. Ref. 8
h. This work
i. Ref. 14
j. Ref. 15

FIGURE CAPTIONS

Fig. 1 Decay chain for A = 140. Half-lives are from Reference 22.

Fig. 2 A probability plot of fractional cumulative yields as a function of Z_H-Z_p.

Fig. 3 A probability plot of fractional cumulative yields from fission of ^{249}Cf and ^{252}Cf adjusted to those from fission of ^{233}U and ^{235}U. Symbols are same as those used in Fig. 2.

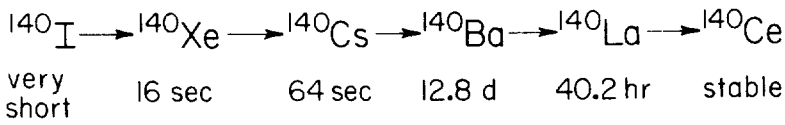

FIG. 1 Decay chain for A = 140. Half-lives are from Reference 22.

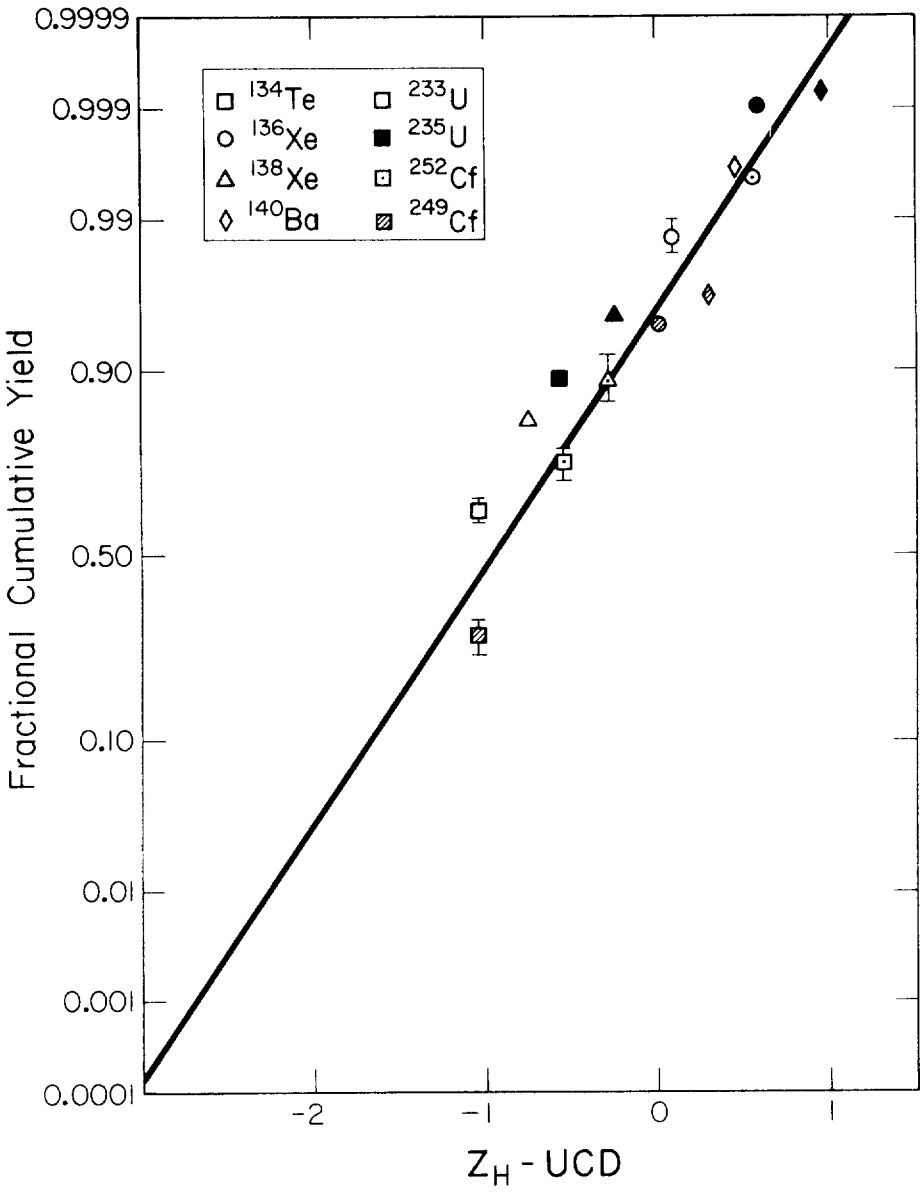

FIG. 2 A probability plot of fractional cumulative yields as a function of Z_H-Z_p.

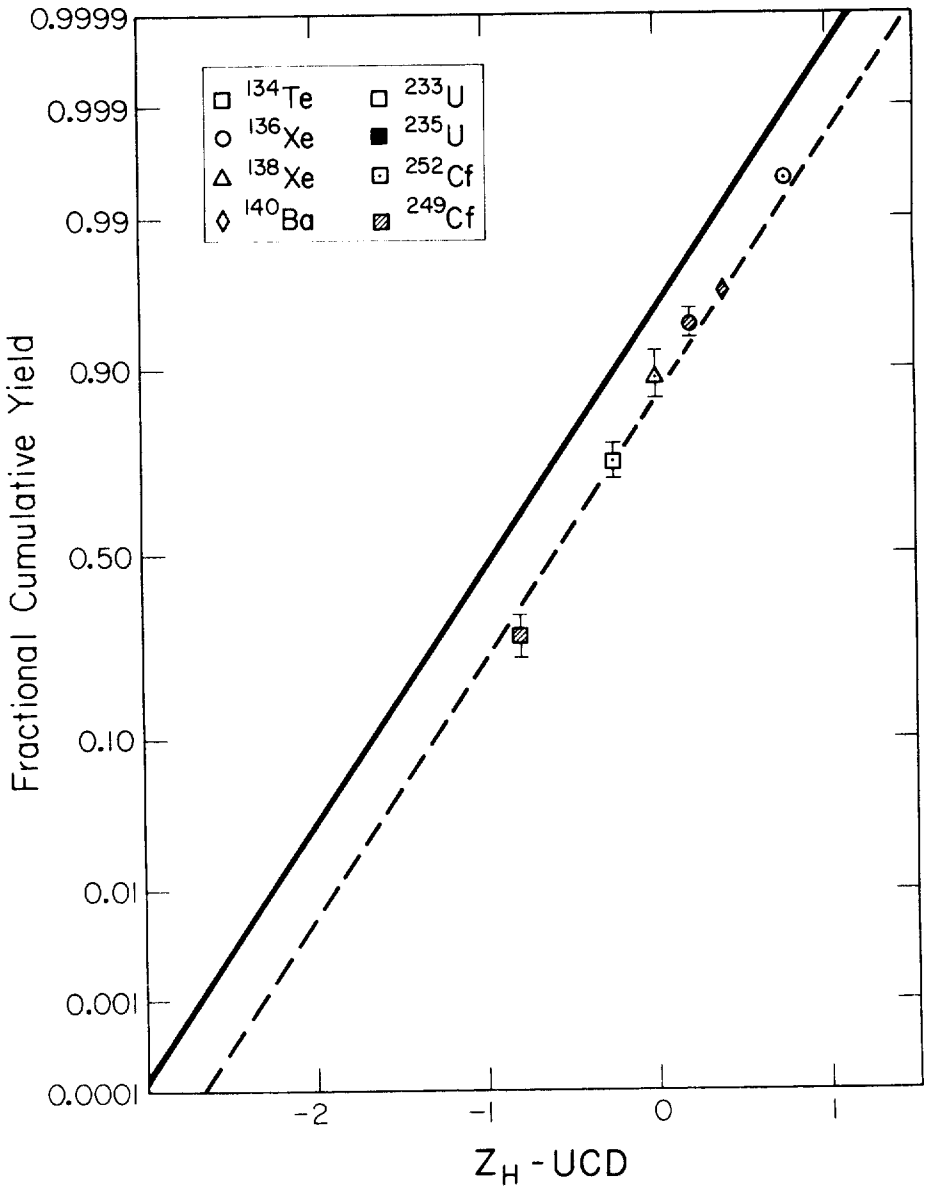

FIG. 3 A probability plot of fractional cumulative yields from fission of ^{249}Cf and ^{252}Cf normalized to those from fission of ^{233}U and ^{235}U. Symbols are same as those used in Fig. 2.